



# Recoil-induced resonances in pump-probe spectroscopy including effects of level degeneracy

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Theoretical calculations are presented for the probe absorption spectra of atoms having magnetically degenerate ground states which are subjected to weak pump fields that produce spatial polarization gradients. Apart from the Raman resonance contribution to the spectrum which has been observed experimentally, we predict that additional resonant structures should appear as a result of the atomic recoil that occurs during the absorption or emission of radiation by the atoms. The width of the atomic recoil resonances is directly related to the Doppler width associated with the driven transition. For sub-Doppler cooled atoms, the width can be much narrower than the optical pumping rate that determines the width of the Raman signal.

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## I. INTRODUCTION

The field of sub-Doppler laser cooling of neutral atoms has progressed to such a level that it is now possible to obtain atomic vapors at very high density ( $\sim 10^{10} \text{ cm}^{-3}$ ) and extremely low temperature ( $\sim 2 \mu\text{K}$ ) [1]. Since the Doppler and transit broadening are greatly reduced in these systems, such cold and dense samples provide ideal environments for carrying out many nonlinear spectroscopic experiments. Moreover, one might expect some new and interesting results which are absent or nonobservable at normal temperatures. For example, the width of the velocity distribution of atoms at room temperature is much larger than the photon recoil velocity  $\hbar k/m$ , where  $k$  is the wave vector of light and  $m$  is the atomic mass. As a result, effects associated with changes in atomic velocity upon absorption or emission of a photon can generally be neglected when considering the spectroscopy of thermal atoms. However, when the distribution width itself is of the order of several  $\hbar k/m$  as in the case of sub-Doppler temperature atoms, such recoil effects may not be negligible. It has already been shown that the inclusion of recoil effects leads to new resonance structures in the nonlinear spectroscopy of an ensemble of "two-level" atoms cooled to sub-Doppler temperatures [2]. The resonances are centered at  $\delta=0$ , where  $\delta$  is the frequency detuning of a probe field from a pump-field frequency. The width of the resonances is characterized by the Doppler width of the ensemble  $ku = kp_0/m$ , where  $u$  is the most probable atomic speed and  $p_0 = mu$  characterizes the momentum distribution width. For many atom-field interactions, the two-level approximation is not satisfactory. This is especially true in sub-Doppler laser cooling where the magnetic degeneracy of the ground-state levels plays a central role in the cooling process.

In this paper, we extend our previous calculations to allow for the magnetic degeneracy of the atomic levels. The inclusion of magnetic-state degeneracy introduces a new dimension in the problem. In both pump-probe and four-wave mixing spectroscopy involving magnetically degenerate atoms, it is possible to observe narrow reso-

nances centered at  $\delta=0$ , whose width is characterized by the rate  $\Gamma'$  at which the ground-state sublevels are optically pumped by the fields [3]. It is not obvious that the recoil-induced resonances, characterized by a width  $ku$  assumed to be much smaller than  $\Gamma'$ , are not obliterated by the optical pumping resonances. It turns out, however, that the recoil-induced resonances persist in the presence of optical pumping and lead to narrow resonant structures superimposed on the line shapes obtained when recoil effects are neglected. Such narrow resonances may have been already observed experimentally [4].

To be specific, we consider the absorption of a probe field having frequency  $\Omega+\delta$  in the presence of a pump field having frequency  $\Omega$ . Generally speaking, the probe absorption is proportional to ground-state density-matrix elements. In addition to the ground-state population, the absorption also depends on differences in population among different magnetic sublevels, as well as magnetic-state coherence. Such quantities are conveniently expressed in an irreducible tensor basis as  $\rho_0^0(g)$ ,  $\rho_Q^1(g)$ , and  $\rho_{Q+1,+2}^2(g)$ , where  $\rho_0^0(g)$  is proportional to the ground-state total population, and  $\rho_Q^1(g)$  and  $\rho_Q^2(g)$  represent ground-state orientation and alignment, respectively. Under the low-field-intensity approximation to be defined below, one can adiabatically eliminate the excited-state quantities and obtain a set of equations involving only ground-state density-matrix elements [3,5]. As a direct result of these equations,  $\rho_0^0(g)$  remains constant in the absence of recoil while  $\rho_Q^K(g)$  ( $K \neq 0$ ) decay at rates characterized by the optical pumping rate  $\Gamma' = (\Gamma|\chi|^2)/[(\Gamma/2)^2 + \Delta^2]$ , where  $\chi$  is a pump-field Rabi frequency,  $\Gamma$  is the excited-state population decay rate, and  $\Delta$  is the pump detuning from the atomic resonance frequency. As a consequence, one might expect that the nonlinear pump-probe signals should have a width at least of order  $\Gamma'$  in the absence of recoil or some other motional narrowing effects.

To take into account recoil effect, one needs to quantize the atomic center-of-mass momentum and write the density-matrix elements as  $\rho(\mathbf{p}, \mathbf{p}')$ . As a result of the

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recoil shifts, the momentum-integrated off-diagonal matrix elements  $\int \rho_0^0(\mathbf{p}, \mathbf{p} + \Delta\mathbf{p}) d\mathbf{p}$  for  $\Delta\mathbf{p} \neq 0$  is not conserved. Assuming the initial  $\rho_0^0(\mathbf{p}, \mathbf{p}')$  is diagonal in  $\mathbf{p}$  space, then owing to the interaction of the atoms with the pump and probe fields, off-diagonal elements of  $\rho_0^0(\mathbf{p}, \mathbf{p}')$  are generated, which are responsible for the recoil-induced signals.

The recoil-induced probe absorption or amplification is most easily understood in terms of Raman transitions between atomic center-of-mass momentum states, as mentioned at the end of Ref. [2]. We want to show here in some more detail that there exists a close analogy between Raman process between internal atomic states and Raman process between external momentum states. As an example of the former, consider the three-level system shown in Fig. 1(a). The two ground states  $|a\rangle$  and  $|b\rangle$  are separated by some finite-energy splitting  $\delta E_{ha} > 0$ . The frequencies of the pump and probe fields are given by  $\Omega$  and  $\Omega' = \Omega + \delta$ , respectively. Both frequencies are assumed to be detuned far from the resonance frequency between state  $|a\rangle$  or  $|b\rangle$  and the excited state  $|c\rangle$ ; therefore, the population of state  $|c\rangle$  is negligible, and states  $|a\rangle$  and  $|b\rangle$  are connected by stimulated transitions involving both pump and probe fields. Assume that  $\rho_a > \rho_b$ , where  $\rho_a$  and  $\rho_b$  are the populations of states  $|a\rangle$  and  $|b\rangle$ . For  $\delta > 0$ , there is more absorption of probe photons and emission of pump photons than the other way around; as a result one has a net probe loss [see Fig. 1(a)]. By the same token, for  $\delta < 0$ , one has a net probe gain.

Now consider the case of a Raman-type transition between the atomic momentum states. Assume that the atomic system has some momentum distribution centered at  $\mathbf{p}=0$  whose width is on the order of several  $\hbar k$ , as

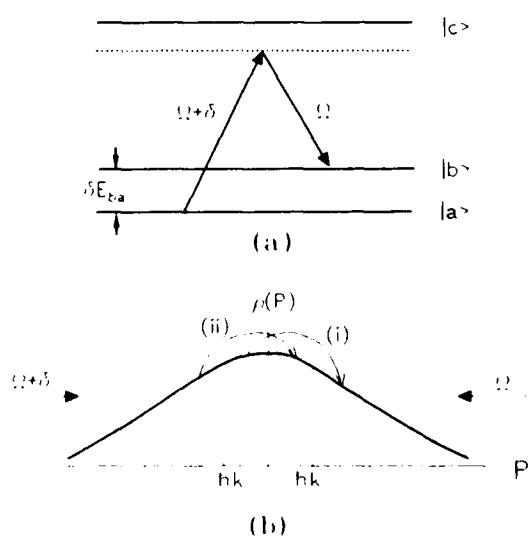


FIG. 1. (a) Energy diagrams for a three-level atom. The Raman transitions are between the two ground states  $|a\rangle$  and  $|b\rangle$ , whose energies are separated by an amount  $\delta E_{ha}$ . (b) The center-of-mass momentum distribution of atoms. Raman transitions occur between momentum states  $p$  and  $p + 2\hbar k$  via absorption of a probe photon and emission of a pump photon. Based on conservation of energy, arrow (i) corresponds to  $\delta > 0$  ( $p > -\hbar k$ ), and arrow (ii) to  $\delta < 0$  ( $p < -\hbar k$ ).

shown in Fig. 1(b). The probe and pump propagate in the  $+\mathbf{k}$  and  $-\mathbf{k}$  directions, respectively. If the initial atomic momentum in the  $\mathbf{k}$  direction is  $p$ , then after absorption of one probe photon and emission of one pump photon, the final-state momentum is  $p + 2\hbar k$ . The resonance condition of such a process obtained from conservation of energy involving the center-of-mass degree of freedom is given by

$$\delta - \frac{2kp}{m} - 4\omega_k = 0, \quad (1)$$

where  $\omega_k = \hbar k^2/2m$  is the recoil frequency. Assuming that the momentum distribution remains unchanged during the course of interaction, the resonance width is essentially determined by the interaction time with the fields. When  $\delta > 0$ , the initial momentum obtained from Eq. (1) satisfies  $p > -\hbar k$ ; consequently, the final momentum is  $(p + 2\hbar k) > \hbar k$ . As one can see, the initial momentum state is more populated than the final momentum state, resulting in a net probe loss. Similarly, when  $\delta < 0$ , the initial momentum state is less populated than the final momentum state and one has a net probe gain. When  $\delta = 0$ , initial and final momentum state have the same population; there is no net probe gain or absorption. One can analyze in the same way the other process of going from  $p$  to  $p - 2\hbar k$  by absorption of one pump photon and emission of one probe photon and it leads to the same conclusion as above. The whole effects are related to the difference of populations between two momentum states differing by  $2\hbar k$ , therefore one would expect that these effects will become significant for the signal generation only when the atomic momentum distribution width is of the order of several  $\hbar k$ . Also from the above analysis, it's easy to see that the signal has a width of order  $ku$  as one tunes  $\delta$  across the momentum distribution. The magnitude of the recoil-induced signal is proportional to the quantity  $\hbar k / (p_0 ku) \sim \omega_k / (ku)^2$ . Since the magnitudes of the background signals having width of order  $\Gamma'$  are proportional to  $1/\Gamma'$ , the dimensionless parameter that determines the amplitude of the recoil-induced signal to the resonance having width  $\Gamma'$  is of order

$$\frac{\Gamma' \hbar k}{ku} \sim \frac{\Gamma' \omega_k}{(ku)^2}. \quad (2)$$

As will be discussed below, this quantity can be comparable to unity.

As a specific example, we consider the case of a  $J_g = 1 \rightarrow J_e = 2$  transition driven by two pump fields having  $\sigma^+$  and  $\sigma^-$  polarizations and a  $\sigma^+$  probe field. For this type of pump-field configuration, the atomic distribution function is uniform in space, owing to the fact that the energy shifts of the atomic ground-state sublevels are space independent [3]. Therefore, any spatial localization effects of the atoms can be neglected. These effects can be important for some other situations, such as with two perpendicularly polarized pump fields, and they can introduce further complications into the calculations. We shall address these other pump-field configurations at the end of the article. This paper is arranged as follows. In Sec. II, we derive the generalized optical Bloch equations [6] for the atomic density-matrix elements, and invoke

the low-field-intensity and low-energy approximations. Then, in Sec. III, we calculate the probe absorption signal, which includes the contribution of the recoil-induced resonances between the atomic center-of-mass momentum states, and that of the Raman resonances between the atomic ground-state sublevels. Finally, in Sec. IV, a discussion of our results and some other situations with different pump-field configurations is given.

## II. GENERALIZED OPTICAL BLOCH EQUATIONS

The atomic level scheme, along with the relative strengths of the transitions (Clebsch-Gordan coefficients associated with the transitions) is shown in Fig. 2(a). The ground and excited states have angular momenta  $J_g = 1$  and  $J_e = 2$ , respectively. The incident-field configuration, shown in Fig. 2(b), is as follows: the two pumps fields propagate in the  $\pm \hat{z}$  directions, having  $\sigma^+$  and  $\sigma^-$  polarizations, respectively. The probe field is  $\sigma^-$  polarized, propagating in a direction opposite to that of the  $\sigma^+$  pump. The total field can be written as

$$\mathbf{E} = \frac{1}{2} (\mathcal{E}_+ \epsilon_+ e^{ikz-i\Omega t} + \mathcal{E}_- \epsilon_- e^{-ikz-i\Omega t} + \mathcal{E}' \epsilon_- e^{ikz-i(\Omega+\delta)t}) + \text{c.c.}, \quad (3)$$

where  $\epsilon_{\pm} = \mp(1/\sqrt{2})(\hat{x} \pm i\hat{y})$ , and  $\hat{x}$  and  $\hat{y}$  are unit vectors in  $\hat{x}$  and  $\hat{y}$  directions, respectively. This pump-field configuration is known to lead to sub-Doppler cooling [3].

The Hamiltonian for the atomic system including the center-of-mass motion is given by

$$H = \frac{\mathbf{P}^2}{2m} + \hbar\omega \sum_{m=-2}^2 |\langle em \rangle \langle em | + V, \quad (4)$$

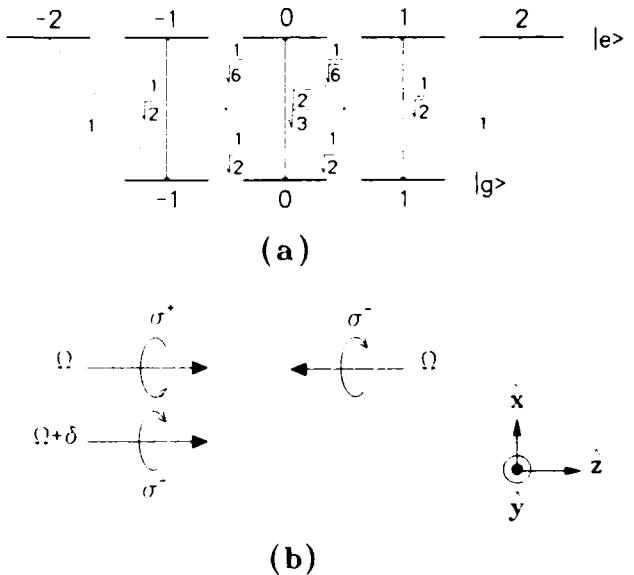


FIG. 2. (a) The atomic level scheme of a  $J_g = 1 \rightarrow J_e = 2$  transition, along with the Clebsch-Gordan coefficients associated with various transitions. (b) The incident-field configuration for the  $J_g = 1 \rightarrow J_e = 2$  transition. The forward and backward pump fields, both having frequency  $\Omega$ , are  $\sigma^+$  and  $\sigma^-$  polarized, respectively. A probe field of frequency  $\Omega + \delta$  is  $\sigma^-$  polarized and propagates in the same direction as the  $\sigma^+$  pump.

where  $p$  is the  $z$  component of the atomic center-of-mass momentum, and  $\omega$  is the atomic resonance frequency. In the rotating-wave approximation, the interaction Hamiltonian  $V$  in the combined basis of magnetic substates  $|g, m\rangle$  ( $m = -1, \dots, 1$ ) and  $|e, m'\rangle$  ( $m' = -2, \dots, 2$ ), and the atomic center-of-mass momentum state  $|p\rangle$  can be written as

$$\begin{aligned} V = & \sum_p \hbar \chi_{+,e} e^{-i\Omega t} \left[ |e2,p+\hbar k\rangle \langle g1,p| + \frac{1}{\sqrt{2}} |e1,p+\hbar k\rangle \langle g0,p| + \frac{1}{\sqrt{6}} |e0,p+\hbar k\rangle \langle g-1,p| \right] \\ & + \sum_p [\hbar \chi_{-,e} e^{-i\Omega t} |e-2,p-\hbar k\rangle \langle g-1,p| + \hbar \chi'_{-e} e^{-i(\Omega+\delta)t} |e-2,p+\hbar k\rangle \langle g-1,p| \\ & + \frac{1}{\sqrt{2}} (\hbar \chi_{-,e} e^{-i\Omega t} |e-1,p-\hbar k\rangle \langle g0,p| + \hbar \chi'_{-e} e^{-i(\Omega+\delta)t} |e-1,p+\hbar k\rangle \langle g0,p|) \\ & + \frac{1}{\sqrt{6}} (\hbar \chi_{-,e} e^{-i\Omega t} |e0,p-\hbar k\rangle \langle g1,p| + \hbar \chi'_{-e} e^{-i(\Omega+\delta)t} |e0,p+\hbar k\rangle \langle g1,p|)] + \text{H.c.}, \end{aligned} \quad (5)$$

where

$$\chi_{\pm} = \frac{er_{eg}\delta_{\pm}}{2\sqrt{5}\hbar}, \quad \chi'_{\pm} = \frac{er_{eg}\delta'_{\pm}}{2\sqrt{5}\hbar}, \quad (6)$$

and  $r_{eg}$  is a reduced matrix element of the dipole moment operator. In deriving Eq. (5), the property

$$e^{+ikz}|p\rangle = |p + \hbar k\rangle \quad (7)$$

of the atomic center-of-mass position operator  $z$  has been used. The atomic density matrix is expanded as

$$\hat{\rho} = \sum_{\alpha,\beta} \rho_{\alpha,\beta}(p,p') |\alpha,p\rangle \langle \beta,p'|, \quad (8)$$

where  $\alpha, \beta = |g, m\rangle, |e, m'\rangle$ . Introducing an interaction representation defined by

$$\rho_{\alpha,\alpha}(p,p') = \tilde{\rho}_{\alpha,\alpha}(p,p'), \quad (9)$$

$$\rho_{gm,em'}(p,p') = \tilde{\rho}_{gm,em'}(p,p') e^{i\omega t},$$

one can use the Schrödinger equation with the Hamiltonian (4) to obtain a set of generalized optical Bloch equations for  $\tilde{\rho}_{\alpha,\beta}(p,p')$ . For example, one finds

$$\dot{\bar{\rho}}_{g-1,e-2}(p,p') = - \left[ \gamma + i \frac{p^2 - p'^2}{2m\hbar} \right] \bar{\rho}_{g-1,e-2}(p,p') - i \chi^* e^{i\Delta t} [\bar{\rho}_{e-2,e-2}(p-\hbar k, p') - \bar{\rho}_{g-1,g-1}(p, p' + \hbar k)] - i \chi'^* e^{i\Delta t} [\bar{\rho}_{e-2,e-2}(p + \hbar k, p') - \bar{\rho}_{g-1,g-1}(p, p' - \hbar k)], \quad (10)$$

where

$$\gamma = \Gamma / 2$$

is the electronic state coherence decay rate, and

$$\Delta = \Omega - \omega, \quad \Delta' = \Omega' - \omega = \Delta + \delta. \quad (11)$$

We now invoke a low-field-intensity and a low-energy approximation [6]. In the low-field-intensity limit

$$|\chi_{\pm}| \ll \Gamma, |\Delta|, \quad (12)$$

quasi-steady-state solutions for the electronic state coherence and excited state density-matrix elements can be obtained in terms of ground-state density-matrix elements. For example, the approximate solution of Eq. (10) is given by

$$\begin{aligned} \bar{\rho}_{g-1,e-2}(p,p') = & \frac{i \chi^* e^{i\Delta t}}{\gamma + i \left[ \Delta + \frac{p^2 - p'^2}{2m\hbar} \right]} \\ & \times \bar{\rho}_{g-1,g-1}(p, p' + \hbar k) \\ & + \frac{i \chi'^* e^{i\Delta t}}{\gamma + i \left[ \Delta' + \frac{p^2 - p'^2}{2m\hbar} \right]} \\ & \times \bar{\rho}_{g-1,g-1}(p, p' - \hbar k). \end{aligned} \quad (13)$$

When substituting similar expressions for  $\bar{\rho}_{ge}$ ,  $\rho_{eg}$ , and  $\bar{\rho}_{ee}$  into the evolution equations for ground-state quantities, one obtains a set of equations involving ground-state density-matrix elements only. Furthermore, we assume the atomic center-of-mass energy is sufficiently small to allow us to neglect the kinetic-energy term  $(p^2 - p'^2)/2m\hbar$  in comparison with  $|\Delta|$  (or  $|\Delta'|$ ). Both the low-field-intensity and low-energy approximations are generally valid in regimes appropriate to sub-Doppler cooling, assuming, as we do in this paper, that  $|\Delta| \geq \Gamma$ . With these two approximations, one obtains a set of simplified Bloch equations for the atomic ground-state elements. They are given in the next section.

Using the low-field-intensity and low-energy approximations, we solve for the probe absorption coefficient to all orders in the pump-field strength and to first order in the probe-field strength. As shown in Appendix A, the pump-field modified probe absorption is related to an atomic coherence  $\bar{\rho}_{ge}$ , which can be expressed in terms of the ground-state density-matrix elements as

$$\begin{aligned} \bar{\rho}_{ge} = & \frac{5}{9} \frac{i \chi^*}{\gamma + i \Delta'} \int \rho_s^{(1)}(p, p + 2\hbar k) dp \\ & - \frac{5\sqrt{2}}{12} \frac{i \chi^*}{\gamma + i \Delta} \left[ \rho_{\text{or}}^{(1)} - \frac{1}{5\sqrt{3}} \rho_{\text{al}}^{(1)} - \frac{\sqrt{2}}{5} \rho_{1,-1}^{(1)} \right], \end{aligned} \quad (14)$$

where we have assumed that  $\chi_+ = \chi_- = \chi$ , and the quantities on the right-hand side of the above equation are defined by

$$\rho_s^{(1)}(p, p + 2\hbar k) = \bar{\rho}_s^{(1)}(p, p + 2\hbar k) e^{-i\delta_i}, \quad (15)$$

where

$$\bar{\rho}_s(p, p') = \sum_{m=-1}^1 \bar{\rho}_{gm, gm}(p, p'), \quad (16)$$

and

$$\begin{aligned} \rho_{\text{or}}^{(1)} &= e^{-i\delta_i} \int \bar{\rho}_{\text{or}}^{(1)}(p, p + 2\hbar k) dp, \\ \rho_{\text{al}}^{(1)} &= e^{-i\delta_i} \int \bar{\rho}_{\text{al}}^{(1)}(p, p + 2\hbar k) dp, \\ \rho_{1,-1}^{(1)} &= e^{-i\delta_i} \int \bar{\rho}_{1,-1}^{(1)}(p, p) dp, \end{aligned} \quad (17)$$

where

$$\begin{aligned} \bar{\rho}_{\text{or}}(p, p') &= \frac{1}{\sqrt{2}} [\bar{\rho}_{g1,g1}(p, p') - \bar{\rho}_{g-1,g-1}(p, p')], \\ \bar{\rho}_{\text{al}}(p, p') &= \frac{1}{\sqrt{6}} [\bar{\rho}_{g1,g1}(p, p') + \bar{\rho}_{g-1,g-1}(p, p') \\ &\quad - 2\bar{\rho}_{g0,g0}(p, p')]. \end{aligned} \quad (18)$$

The superscript (1) of various  $\rho$ 's or  $\bar{\rho}$ 's in Eqs. (15) and (17) indicates that these quantities are proportional to  $\chi^*$  (in the resonance or rotating-wave approximation, there is no contribution to  $\bar{\rho}_{ge}$  which is proportional to  $\chi'$ ). Also in this paper, unless specified otherwise, a superscript (0) of a density-matrix element indicates that this element is zeroth order in probe-field strength, while a density-matrix element without any superscript means that it is both zeroth and first order in probe strength.

Before going into the details of the calculation, it is helpful to identify various terms in Eq. (14) with different physical processes that contribute to the signal. The first term,  $\rho_s^{(1)}$ , represents the signal that is related to the probe-induced spatial modulation of the total atomic population. Such a contribution has not been considered before in similar contexts. As we will see below, it is a signal due to the recoil-induced resonances involving both the probe and the pump fields. The term  $\rho_{\text{or}}^{(1)}$  determines the nonvanishing component of the ground-state orientation, while  $\rho_{\text{al}}^{(1)}$  and  $\rho_{1,-1}^{(1)}$  determine different components of the atomic ground-state alignment. In the limit that  $|\Delta| \gg \Gamma$ , they represent the signal due to Raman processes between the atomic ground-state sublevels. Both contributions are analyzed in the next section.

### III. CALCULATION OF THE PROBE ABSORPTION SIGNAL

In this section, we derive in detail the probe absorption coefficient given by Eq. (14). Our calculation is based on

the generalized optical Bloch equations described above. In the low-field-intensity and low-energy approximations, and keeping terms to first order in  $\chi'^*$  only (and dropping

terms proportional to  $\chi'$  which do not contribute to  $\tilde{\rho}_{ge}$  to linear order in probe-field strength), the equations for the ground-state density-matrix elements are given by

$$\begin{aligned}
 \dot{\tilde{\rho}}_{g-1,g-1}(p,p') = & - \left[ \gamma_g + i \frac{p^2 - p'^2}{2m\hbar} \right] \tilde{\rho}_{g-1,g-1}(p,p') + \gamma_g \tilde{\rho}_{g-1,g-1}^{(0)}(p,p') - \frac{\gamma}{6} \Gamma' \tilde{\rho}_{g-1,g-1}(p,p') \\
 & - \frac{\chi^2}{6(\gamma - i\Delta)} \tilde{\rho}_{g1,g-1}(p+2\hbar k, p') - \frac{\chi^2}{6(\gamma + i\Delta)} \tilde{\rho}_{g-1,g1}(p, p'+2\hbar k) \\
 & + \Gamma' \int dq N_-(q) \tilde{\rho}_{g-1,g-1}(p+\hbar k + \hbar q, p'+\hbar k + \hbar q) \\
 & + \frac{\Gamma'}{4} \int dq N_0(q) \tilde{\rho}_{g0,g0}(p+\hbar k + \hbar q, p'+\hbar k + \hbar q) \\
 & + \frac{\Gamma'}{36} \int dq N_+(q) [\tilde{\rho}_{g-1,g-1}(p-\hbar k + \hbar q, p'-\hbar k + \hbar q) + \tilde{\rho}_{g1,g1}(p+\hbar k + \hbar q, p'+\hbar k + \hbar q) \\
 & \quad + \tilde{\rho}_{g-1,g1}(p-\hbar k + \hbar q, p'+\hbar k + \hbar q) + \tilde{\rho}_{g1,g-1}(p+\hbar k + \hbar q, p'-\hbar k + \hbar q)] \\
 & - \frac{\chi \chi'^* e^{i\delta t}}{\gamma - i\Delta} \tilde{\rho}_{g-1,g-1}(p+2\hbar k, p') - \frac{\chi \chi'^* e^{i\delta t}}{\gamma + i\Delta'} \tilde{\rho}_{g-1,g-1}(p, p-2\hbar k) \\
 & + \frac{\Gamma \chi \chi'^* e^{i\delta t}}{\gamma^2 + \Delta^2} \int dq N_-(q) \tilde{\rho}_{g-1,g-1}(p+\hbar k + \hbar q, p-\hbar k + \hbar q) - \frac{\chi \chi'^* e^{i\delta t}}{6(\gamma + i\Delta')} \tilde{\rho}_{g-1,g1}(p, p') \\
 & + \frac{\Gamma \chi \chi'^* e^{i\delta t}}{4(\gamma^2 + \Delta^2)} \int dq N_0(q) \tilde{\rho}_{g0,g0}(p+\hbar k + \hbar q, p'-\hbar k + \hbar q) \\
 & + \frac{\Gamma \chi \chi'^* e^{i\delta t}}{36(\gamma^2 + \Delta^2)} \int dq N_+(q) [\tilde{\rho}_{g1,g1}(p+\hbar k + \hbar q, p'-\hbar k + \hbar q) + \tilde{\rho}_{g-1,g1}(p-\hbar k + \hbar q, p'-\hbar k + \hbar q)], 
 \end{aligned} \tag{19}$$

$$\begin{aligned}
 \dot{\tilde{\rho}}_{g0,g0}(p,p') = & - \left[ \gamma_g + i \frac{p^2 - p'^2}{2m\hbar} \right] \tilde{\rho}_{g0,g0}(p,p') + \gamma_g \tilde{\rho}_{g0,g0}^{(0)}(p,p') - \Gamma' \tilde{\rho}_{g0,g0}(p,p') \\
 & + \frac{\Gamma'}{4} \left[ \int dq N_-(q) \tilde{\rho}_{g0,g0}(p+\hbar k + \hbar q, p'+\hbar k + \hbar q) + \int dq N_+(q) \tilde{\rho}_{g0,g0}(p-\hbar k + \hbar q, p'-\hbar k + \hbar q) \right] \\
 & + \frac{\Gamma'}{9} \int dq N_0(q) [\tilde{\rho}_{g-1,g-1}(p-\hbar k + \hbar q, p'-\hbar k + \hbar q) + \tilde{\rho}_{g1,g1}(p+\hbar k + \hbar q, p'+\hbar k + \hbar q) \\
 & \quad + \tilde{\rho}_{g-1,g1}(p-\hbar k + \hbar q, p'+\hbar k + \hbar q) + \tilde{\rho}_{g1,g-1}(p+\hbar k + \hbar q, p'-\hbar k + \hbar q)] \\
 & - \frac{\chi \chi'^* e^{i\delta t}}{2(\gamma - i\Delta)} \tilde{\rho}_{g0,g0}(p+2\hbar k, p') - \frac{\chi \chi'^* e^{i\delta t}}{2(\gamma + i\Delta')} \tilde{\rho}_{g0,g0}(p, p'-2\hbar k) \\
 & + \frac{\Gamma \chi \chi'^* e^{i\delta t}}{4(\gamma^2 + \Delta^2)} \int dq N_-(q) \tilde{\rho}_{g0,g0}(p+\hbar k + \hbar q, p'-\hbar k + \hbar q) \\
 & + \frac{\Gamma \chi \chi'^* e^{i\delta t}}{9(\gamma^2 + \Delta^2)} \int dq N_0(q) [\tilde{\rho}_{g1,g1}(p+\hbar k + \hbar q, p'-\hbar k + \hbar q) + \tilde{\rho}_{g-1,g1}(p-\hbar k + \hbar q, p'-\hbar k + \hbar q)], 
 \end{aligned} \tag{20}$$

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$$\begin{aligned}
\dot{\tilde{\rho}}_{g1,g1}(p, p') = & - \left[ \gamma_g + i \frac{p^2 - p'^2}{2m\hbar} \right] \tilde{\rho}_{g1,g1}(p, p') + \gamma_g \tilde{\rho}_{g1,g1}^{(0)}(p, p') - \frac{1}{6} \Gamma' \tilde{\rho}_{g1,g1}(p, p') \\
& - \frac{\chi^2}{6(\gamma - i\Delta)} \tilde{\rho}_{g-1,g1}(p - 2\hbar k, p') - \frac{\chi^2}{6(\gamma + i\Delta)} \tilde{\rho}_{g1,g-1}(p, p' - 2\hbar k) \\
& + \Gamma' \int dq N_+(q) \tilde{\rho}_{g1,g1}(p - \hbar k + \hbar q, p' - \hbar k + \hbar q) + \frac{\Gamma'}{4} \int dq N_0(q) \tilde{\rho}_{g0,g0}(p - \hbar k + \hbar q, p' - \hbar k + \hbar q) \\
& + \frac{\Gamma'}{36} \int dq N_-(q) [\tilde{\rho}_{g-1,g-1}(p - \hbar k + \hbar q, p' - \hbar k + \hbar q) + \tilde{\rho}_{g1,g1}(p + \hbar k + \hbar q, p' + \hbar k + \hbar q) \\
& + \tilde{\rho}_{g-1,g1}(p - \hbar k + \hbar q, p' + \hbar k + \hbar q) + \tilde{\rho}_{g1,g-1}(p + \hbar k + \hbar q, p' - \hbar k + \hbar q)] \\
& - \frac{\chi\chi'^* e^{i\delta t}}{6(\gamma - i\Delta)} \tilde{\rho}_{g1,g1}(p + 2\hbar k, p') - \frac{\chi\chi'^* e^{i\delta t}}{6(\gamma + i\Delta')} \tilde{\rho}_{g1,g1}(p, p' - 2\hbar k) \\
& + \frac{\Gamma\chi\chi'^* e^{i\delta t}}{36(\gamma^2 + \Delta^2)} \int dq N_-(q) [\tilde{\rho}_{g1,g1}(p + \hbar k + \hbar q, p' - \hbar k + \hbar q) + \tilde{\rho}_{g-1,g1}(p - \hbar k + \hbar q, p' - \hbar k + \hbar q)] , \quad (21)
\end{aligned}$$

$$\begin{aligned}
\dot{\tilde{\rho}}_{g-1,g1}(p, p') = & - \left[ \gamma_g + i \frac{p^2 - p'^2}{2m\hbar} \right] \tilde{\rho}_{g-1,g1}(p, p') + \gamma_g \tilde{\rho}_{g-1,g1}^{(0)}(p, p') - \frac{1}{6} \Gamma' \tilde{\rho}_{g-1,g1}(p, p') \\
& - \frac{\chi^2}{6(\gamma - i\Delta)} \tilde{\rho}_{g1,g1}(p + 2\hbar k, p') - \frac{\chi^2}{6(\gamma + i\Delta)} \tilde{\rho}_{g-1,g-1}(p, p' - 2\hbar k) \\
& + \frac{\Gamma'}{6} \int dq N_-(q) [\tilde{\rho}_{g-1,g-1}(p + \hbar k + \hbar q, p' - \hbar k + \hbar q) + \tilde{\rho}_{g-1,g1}(p + \hbar k + \hbar q, p' + \hbar k + \hbar q)] \\
& + \frac{\Gamma'}{6} \int dq N_+(q) [\tilde{\rho}_{g1,g1}(p + \hbar k + \hbar q, p' - \hbar k + \hbar q) + \tilde{\rho}_{g-1,g1}(p - \hbar k + \hbar q, p' - \hbar k + \hbar q)] \\
& + \frac{\Gamma'}{4} \int dq N_0(q) \tilde{\rho}_{g0,g0}(p + \hbar k + \hbar q, p' - \hbar k + \hbar q) \\
& - \frac{\chi\chi'^* e^{i\delta t}}{\gamma - i\Delta} \tilde{\rho}_{g-1,g1}(p + 2\hbar k, p') - \frac{\chi\chi'^* e^{i\delta t}}{6(\gamma + i\Delta')} \tilde{\rho}_{g-1,g1}(p, p' - 2\hbar k) \\
& + \frac{\Gamma\chi\chi'^* e^{i\delta t}}{6(\gamma^2 + \Delta^2)} \int dq N_-(q) \tilde{\rho}_{g-1,g1}(p + \hbar k + \hbar q, p' - \hbar k + \hbar q) , \quad (22)
\end{aligned}$$

$$\begin{aligned}
\dot{\tilde{\rho}}_{g1,g-1}(p, p') = & - \left[ \gamma_g + i \frac{p^2 - p'^2}{2m\hbar} \right] \tilde{\rho}_{g1,g-1}(p, p') + \gamma_g \tilde{\rho}_{g1,g-1}^{(0)}(p, p') - \frac{1}{6} \Gamma' \tilde{\rho}_{g1,g-1}(p, p') \\
& - \frac{\chi^2}{6(\gamma - i\Delta)} \tilde{\rho}_{g-1,g-1}(p - 2\hbar k, p') - \frac{\chi^2}{6(\gamma + i\Delta)} \tilde{\rho}_{g1,g1}(p, p' + 2\hbar k) \\
& + \frac{\Gamma'}{6} \int dq N_-(q) [\tilde{\rho}_{g-1,g-1}(p - \hbar k + \hbar q, p' + \hbar k + \hbar q) + \tilde{\rho}_{g1,g-1}(p + \hbar k + \hbar q, p' + \hbar k + \hbar q)] \\
& + \frac{\Gamma'}{6} \int dq N_+(q) [\tilde{\rho}_{g1,g1}(p - \hbar k + \hbar q, p' + \hbar k + \hbar q) + \tilde{\rho}_{g1,g-1}(p - \hbar k + \hbar q, p' - \hbar k + \hbar q)] \\
& + \frac{\Gamma'}{4} \int dq N_0(q) \tilde{\rho}_{g0,g0}(p - \hbar k + \hbar q, p' + \hbar k + \hbar q) - \frac{\chi\chi'^* e^{i\delta t}}{6(\gamma - i\Delta)} \tilde{\rho}_{g-1,g-1}(p, p') - \frac{\chi\chi'^* e^{i\delta t}}{6(\gamma + i\Delta')} \tilde{\rho}_{g1,g1}(p, p') \\
& - \frac{\chi\chi'^* e^{i\delta t}}{6(\gamma - i\Delta)} \tilde{\rho}_{g1,g-1}(p + 2\hbar k, p') - \frac{\chi\chi'^* e^{i\delta t}}{6(\gamma + i\Delta')} \tilde{\rho}_{g1,g-1}(p, p' - 2\hbar k) \\
& + \frac{\Gamma\chi\chi'^* e^{i\delta t}}{6(\gamma^2 + \Delta^2)} \int dq N_-(q) [\tilde{\rho}_{g-1,g-1}(p - \hbar k + \hbar q, p' - \hbar k + \hbar q) + \tilde{\rho}_{g1,g-1}(p + \hbar k + \hbar q, p' - \hbar k + \hbar q)] \\
& + \frac{\Gamma\chi\chi'^* e^{i\delta t}}{6(\gamma^2 + \Delta^2)} \int dq N_+(q) \tilde{\rho}_{g1,g1}(p - \hbar k + \hbar q, p' - \hbar k + \hbar q) \\
& + \frac{\Gamma\chi\chi'^* e^{i\delta t}}{4(\gamma^2 + \Delta^2)} \int dq N_0(q) \tilde{\rho}_{g0,g0}(p - \hbar k + \hbar q, p' - \hbar k + \hbar q) . \quad (23)
\end{aligned}$$

In the above equations,  $\gamma_g$  is an effective ground-state decay rate, which could be determined, for example, by transit-time effects, and  $\tilde{\rho}_{gm,gm}^{(0)}$  are the values of  $\tilde{\rho}_{gm,gm}(p,p')$  to zeroth order in probe-field strength. In our calculation, we assume that the pump fields have created an equilibrium state before the addition of the probe field. Therefore  $\tilde{\rho}_{gm,gm}^{(0)}(p,p')$  are the equilibrium values of the atomic density-matrix elements in the presence of the two pump fields only. If the atomic density matrix is diagonal in momentum space before the pump fields are turned on, then it is easy to deduce from the above equations, when  $\chi'=0$ , that the only nonvanishing density matrix elements are given by [3]

$$\tilde{\rho}_{gm,gm}^{(0)}(p,p) = \tilde{\rho}_{gm,gm}^{(0)}(p), \quad m = -1, 0, 1, \quad (24)$$

and

$$\tilde{\rho}_{g\pm 1,g\pm 1}^{(0)}(p \pm \hbar k, p \mp \hbar k) = \tilde{\rho}_{g\pm 1,g\pm 1}^{(0)}(p). \quad (25)$$

Note that the atomic momentum distribution function in the absence of the probe is defined by

$$\tilde{\rho}_s^{(0)}(p,p) = \sum_{m=-1}^1 \tilde{\rho}_{gm,gm}^{(0)}(p,p). \quad (26)$$

In this paper, we assume that the width of atomic momentum distribution, which is of order  $p_0 = mu$ , satisfies the following condition:

$$\omega_k \ll k u = \frac{kp_0}{m} \ll \Gamma'. \quad (27)$$

The above inequality implies that  $p_0 \gg \hbar k$ .

Finally,  $N_{\pm 0}(q)$  in Eqs. (19)–(23) are the probability densities for emission of a spontaneous photon having a certain polarization ( $\sigma^\pm$  or  $\pi$ ) and a  $z$  component of momentum equal to  $\hbar q$ . They are given by [7]

$$N_{\pm 0}(q) = \frac{3}{8k} \left[ 1 + \frac{q^2}{k^2} \right], \quad N_0(q) = \frac{3}{4k} \left[ 1 - \frac{q^2}{k^2} \right]. \quad (28)$$

In the present calculation, the results are insensitive to the detail of the functions  $N_\epsilon(q)$  ( $\epsilon = \pm 0$ ) as long as they satisfy the relation

$$\int q N_\epsilon(q) dq = 0, \quad (29)$$

which they do. In the following, we use a single normalized probability function  $\tilde{N}(q)$ , which also satisfies Eq. (29), in describing the momentum distribution of a spontaneous photon.

### A. Recoil-induced signal

To calculate the recoil-induced signal, one has to obtain the solution for  $\tilde{\rho}_s(p, p + 2\hbar k)$  [see Eq. (14)]. In order to analyze the effects of the pump fields on the signal, it is convenient to use the momentum-family notation introduced in Ref. [3]. For example, the ground states that belong to the same family  $\mathcal{F}(p)$  are  $|gm, p + m\hbar k\rangle$ ,  $m = -1, 0, 1$ . The stimulated processes involving the pump fields do not change the momentum of a given family. However, the addition of the probe field can induce stimulated transitions between different momentum families, a process that leads to the recoil-induced resonances. Later in this paper, we will discuss another situation where the stimulated transitions involving the probe and the pump fields do not change the momentum of a family.

In the momentum-family notation, we define the ground-state density-matrix elements that are related to the recoil-induced signal as

$$\rho_m(p) = \tilde{\rho}_{gm,gm}(p + m\hbar k, p + m\hbar k + 2\hbar k), \quad m = -1, 0, 1 \quad (30)$$

and

$$\rho_{\pm 1, \pm 1}(p) = \tilde{\rho}_{g\pm 1, g\pm 1}(p \pm \hbar k, p \mp \hbar k + 2\hbar k). \quad (31)$$

Furthermore, the ground-state population, and orientation and alignment components are similarly written as

$$\begin{aligned} \rho_s(p) &= \sum_{m=-1}^1 \rho_m(p), \\ \rho_{\text{or}}(p) &= \frac{1}{\sqrt{2}} [\rho_1(p) - \rho_{-1}(p)], \\ \rho_{\text{al}}(p) &= \frac{1}{\sqrt{6}} [\rho_1(p) + \rho_{-1}(p) - 2\rho_0(p)]. \end{aligned} \quad (32)$$

Since the only nonvanishing density-matrix elements to zeroth order are given by Eqs. (24) and (25), the terms in the above definitions are all first order in the probe-field strength. We have omitted the superscript (1) for convenience. The recoil-induced signal is now proportional to the integral of

$$\int \rho_s(p) dp = \int \tilde{\rho}_s^{(1)}(p, p + 2\hbar k) dp,$$

as is evident from Eq. (14). We now proceed to obtain the equation for the evolution of  $\rho_s(p)$ .

First,  $\rho_{-1, -1}(p)$  are obtained from Eqs. (22) and (23) as

$$\begin{aligned} \rho_{-1, -1}(p) &= -\frac{1}{5\Gamma'} \left[ \frac{\chi^2}{\gamma - i\Delta} \rho_1(p) + \frac{\chi^2}{\gamma + i\Delta} \rho_{-1}(p) \right] \\ &\quad + \frac{1}{5} \int dq \tilde{N}(q) [\rho_{-1}(p + \hbar k + \hbar q) + \rho_1(p - \hbar k + \hbar q) + \frac{3}{2}\rho_0(p + \hbar q)] \\ &\quad - \frac{\chi\chi'^* e^{i\delta t}}{5\Gamma'(\gamma + i\Delta')} \tilde{\rho}_{g-1, g1}^{(0)}(p - \hbar k, p + \hbar k) - \frac{6\chi\chi'^* e^{i\delta t}}{5\Gamma'(\gamma - i\Delta)} \tilde{\rho}_{g-1, g1}^{(0)}(p + \hbar k, p + 3\hbar k) \\ &\quad + \frac{\Gamma\chi\chi'^* e^{i\delta t}}{5\Gamma'(\gamma^2 + \Delta^2)} \int dq \tilde{N}(q) \tilde{\rho}_{g-1, g1}^{(0)}(p + \hbar q, p + 2\hbar q + \hbar q), \end{aligned} \quad (33)$$

and

$$\begin{aligned}
\rho_{1,-1}(p) = & -\frac{1}{5\Gamma'} \left[ \frac{\chi^2}{\gamma-i\Delta} \rho_{-1}(p) + \frac{\chi^2}{\gamma+i\Delta} \rho_1(p) \right] \\
& + \frac{1}{5} \int dq \tilde{N}(q) [\rho_{-1}(p+\hbar k + \hbar q) + \rho_1(p-\hbar k + \hbar q) + \frac{3}{2} \rho_0(p + \hbar q)] \\
& - \frac{\chi \chi'^* e^{i\delta t}}{5\Gamma'} \left[ \frac{\tilde{\rho}_{g-1,g-1}^{(0)}(p+\hbar k, p+\hbar k)}{\gamma-i\Delta} + \frac{\tilde{\rho}_{g1,g1}^{(0)}(p+\hbar k, p+\hbar k)}{\gamma+i\Delta} \right. \\
& \left. + \frac{\tilde{\rho}_{g1,g-1}^{(0)}(p+3\hbar k, p+\hbar k)}{\gamma-i\Delta} + \frac{\tilde{\rho}_{g1,g-1}^{(0)}(p+\hbar k, p-\hbar k)}{\gamma+i\Delta} \right] \\
& + \frac{\Gamma \chi \chi'^* e^{i\delta t}}{5\Gamma'(\gamma^2 + \Delta^2)} [\tilde{\rho}_{g1,g1}^{(0)}(p+\hbar q, p+\hbar q) + \tilde{\rho}_{g-1,g-1}^{(0)}(p+\hbar q, p+\hbar q) \\
& + \tilde{\rho}_{g1,g-1}^{(0)}(p+2\hbar k + \hbar q, p+\hbar q) + \frac{3}{2} \tilde{\rho}_{g0,g0}^{(0)}(p+\hbar q, p+\hbar q)] , \quad (34)
\end{aligned}$$

where we have neglected  $\gamma_g$ ,  $\delta$ , and the kinetic-energy term  $(p^2 - p'^2)/2m\hbar$  in comparison with  $\Gamma'$  [see inequality (27)]. Substituting Eqs. (33) and (34) into Eqs. (19)–(21), and, assuming the secular limit

$$\Gamma/|\Delta| \ll 1 , \quad (35)$$

to lowest order in  $\Gamma/|\Delta|$ , one obtains the equation for  $\dot{\rho}_s(p)$  as

$$\begin{aligned}
\dot{\rho}_s(p) = & -\frac{49}{45} \Gamma' \rho_s(p) + \Gamma' \int dq \tilde{N}(q) \{ \frac{58}{135} [\rho_s(p+\hbar k + \hbar q) + \rho_s(p-\hbar k + \hbar q)] \\
& + \frac{1}{270} [\rho_s(p+2\hbar k + \hbar q) + \rho_s(p-2\hbar k + \hbar q)] + \frac{2}{9} \rho_s(p + \hbar q) \} \\
& + \Gamma' \int dq \tilde{N}(q) \left\{ \frac{22\sqrt{2}}{45} [\rho_{\text{or}}(p-\hbar k + \hbar q) - \rho_{\text{or}}(p+\hbar k + \hbar q)] \right. \\
& \left. + \frac{\sqrt{2}}{180} [\rho_{\text{or}}(p-2\hbar k + \hbar q) - \rho_{\text{or}}(p+2\hbar k + \hbar q)] \right\} \\
& - \frac{2\sqrt{6}}{45} \Gamma' \rho_{\text{al}}(p) + \frac{\sqrt{6}}{540} \Gamma' \int dq \tilde{N}(q) \{ \rho_{\text{al}}(p+2\hbar k + \hbar q) + \rho_{\text{al}}(p-2\hbar k + \hbar q) \\
& + 44[\rho_{\text{al}}(p+\hbar k + \hbar q) + \rho_{\text{al}}(p-\hbar k + \hbar q)] - 66 \rho_{\text{al}}(p + \hbar q) \} \\
& - \left[ \gamma_g - i \frac{2kp}{m} - i4\omega_k \right] \rho_s(p) - i4\omega_k \rho_{\text{or}}(p) \\
& - \frac{\chi \chi'^* e^{i\delta t}}{\gamma-i\Delta} \tilde{\rho}_{g-1,g-1}^{(0)}(p+\hbar k, p+\hbar k) - \frac{\chi \chi'^* e^{i\delta t}}{\gamma+i\Delta} \tilde{\rho}_{g-1,g-1}^{(0)}(p-\hbar k, p-\hbar k) \\
& + \frac{\Gamma \chi \chi'^* e^{i\delta t}}{\gamma^2 + \Delta^2} \int dq \tilde{N}(q) \tilde{\rho}_{g-1,g-1}^{(0)}(p+\hbar q, p+\hbar q) \\
& - \frac{\chi \chi'^* e^{i\delta t}}{2(\gamma-i\Delta)} \tilde{\rho}_{g0,g0}^{(0)}(p+2\hbar k, p+2\hbar k) - \frac{\chi \chi'^* e^{i\delta t}}{2(\gamma+i\Delta')} \tilde{\rho}_{g0,g0}^{(0)}(p, p) \\
& + \frac{\Gamma \chi \chi'^* e^{i\delta t}}{2(\gamma^2 + \Delta^2)} \int dq \tilde{N}(q) \tilde{\rho}_{g0,g0}^{(0)}(p+\hbar k + \hbar q, p+\hbar k + \hbar q) \\
& - \frac{\chi \chi'^* e^{i\delta t}}{6(\gamma-i\Delta)} \tilde{\rho}_{g1,g1}^{(0)}(p+3\hbar k, p+3\hbar k) - \frac{\chi \chi'^* e^{i\delta t}}{6(\gamma+i\Delta')} \tilde{\rho}_{g1,g1}^{(0)}(p+\hbar k, p+\hbar k) \\
& + \frac{\Gamma \chi \chi'^* e^{i\delta t}}{6(\gamma^2 + \Delta^2)} \int dq \tilde{N}(q) \tilde{\rho}_{g1,g1}^{(0)}(p+2\hbar k + \hbar q, p+2\hbar k + \hbar q) \\
& - \frac{\chi \chi'^* e^{i\delta t}}{6(\gamma+i\Delta')} \tilde{\rho}_{g-1,g1}^{(0)}(p-\hbar k, p+\hbar k) - \frac{\chi \chi'^* e^{i\delta t}}{6(\gamma-i\Delta)} \tilde{\rho}_{g-1,g1}^{(0)}(p+\hbar k, p+3\hbar k) \\
& - \frac{\Gamma \chi \chi'^* e^{i\delta t}}{6(\gamma^2 + \Delta^2)} \int dq \tilde{N}(q) \tilde{\rho}_{g-1,g1}^{(0)}(p+\hbar q, p+2\hbar k + \hbar q) . \quad (36)
\end{aligned}$$

Note that, neglecting the photon recoil momentum  $\hbar k$  and  $\hbar q$ , one has

$$\rho_s(p) = 0.$$

In other words, when the photon recoil momentum is neglected, the population modulation term  $\rho_s(p)$  remains zero despite the addition of the probe field. This is in agreement with the semiclassical theory where the atomic center-of-mass motion is described by a classical variable (the velocity), and the total atomic population is a constant [5]. In this case the population will remain diagonal in momentum space before and after the addition of the probe field [see Eq. (24)]. It is clear here that the signal related to the probe-induced modification of  $\rho_s(p)$  is a direct result of the photon recoil momentum, which explains why we call this part of contribution to the probe absorption the recoil-induced signal.

From Eqs. (19)–(21), we then solve for the quasi-steady-state solutions for  $\rho_{\text{or}}(p)$  and  $\rho_{\text{al}}(p)$  in terms of  $\rho_s(p)$ , and substitute the solutions into Eq. (36). In the secular limit (35),  $\rho_{\text{or}}(p)$ , whose value is proportional to

$(\Gamma/\Delta)^2(\omega_k/ku)\rho_s(p)$ , can be neglected, while  $\rho_{\text{al}}(p)$  is approximately given by

$$\rho_{\text{al}}(p) = \frac{5\sqrt{6}}{102}\rho_s(p). \quad (37)$$

Corrections to Eq. (37) are smaller by a factor  $\omega_k/ku$ . We have dropped some terms in the solution for  $\rho_{\text{al}}(p)$  that are explicitly proportional to  $\chi'^*$ . Since their contributions to the recoil-induced signal, upon substitution of  $\rho_{\text{al}}(p)$  into Eq. (36), is of order  $\omega_k^2/(ku)^2$  [8], and since we are interested only in the signal that is linear in  $\omega_k/(ku)$  [2], such  $\chi'^*$  terms can be neglected when replacing  $\rho_{\text{al}}(p)$  in Eq. (36) with Eq. (37). After such procedures, we arrive at an equation involving  $\rho_s(p)$  only. In this equation, there are two types of terms. First, there are terms which are explicitly proportional to  $\chi'^*$ . They are identical to those appearing in Eq. (36), and provide the source terms for the recoil-induced signals. Second, there are terms which are proportional to  $\Gamma'$ . They describe the optical pumping effects due to the presence of the pump fields, and are given by

$$[\dot{\rho}_s(p)]_{\text{pump}} = -\frac{281}{255}\Gamma'\rho_s(p) + \frac{13}{3060}\Gamma' \int dq \bar{N}(q)[\rho_s(p+2\hbar k+\hbar q) + \rho_s(p-2\hbar k+\hbar q)] \\ + \frac{347}{765}\Gamma' \int dq N(q)[\rho_s(p+\hbar k+\hbar q) + \rho_s(p-\hbar k+\hbar q)] + \frac{19}{102}\Gamma' \int dq \bar{N}(q)\rho_s(p+\hbar q). \quad (38)$$

If one assumes that  $\bar{N}(q)=\delta(q)$  and expands the integrands around  $p$ , it can be simplified as

$$[\dot{\rho}_s(p)]_{\text{pump}} = \frac{8}{17}\Gamma'(\hbar k)^2 \frac{\partial^2}{\partial p^2} \rho_s(p), \quad (39)$$

where the overdot symbol in Eq. (39) denotes time derivative only. Equation (39) describes a momentum diffusion process induced by the pump fields in the absence of the cooling forces (since we have neglected the Doppler shift in comparison with  $\Gamma'$ ). The diffusion coefficient,  $\frac{8}{17}\Gamma'(\hbar k)^2$ , is in agreement with the result of Ref. [3] in the limit  $|\Delta| \gg \Gamma$  and neglecting the contribution of spontaneous emissions.

In this paper, we are interested only in the qualitative effects of the optical pumping on the recoil-induced sig-

nal. For the sake of simplicity, and without loss of the basic features involved, we replace the optical pumping terms in Eq. (38) by the following simplified model:

$$[\dot{\rho}_s(p)]_{\text{pump}} = -\Gamma_p[2\rho_s(p) - \rho_s(p+\hbar k) - \rho_s(p-\hbar k)], \quad (40)$$

where  $\Gamma_p(\hbar k)^2$  is the atomic momentum diffusion coefficient due to the pump fields, and  $\Gamma_p$  is of order  $\Gamma'$ . One can verify that, by expanding the  $\rho_s(p \pm \hbar k)$  terms in the above equation around  $p$ , Eq. (40) leads to an equation for momentum diffusion that has the same structure as Eq. (39). The complete equation for  $\dot{\rho}_s(p)$  can now be written as

$$\dot{\rho}_s(p) = -\Gamma_p[2\rho_s(p) - \rho_s(p+\hbar k) - \rho_s(p-\hbar k)] - \left[ \gamma_g - i\frac{2kp}{m} - i4\omega_k \right] \rho_s(p) \\ - \frac{\chi\chi'^* e^{i\delta t}}{\gamma - i\Delta} \tilde{\rho}_{g1,g-1}^{(0)}(p+\hbar k, p+\hbar k) - \frac{\chi\chi'^* e^{i\delta t}}{\gamma + i\Delta} \tilde{\rho}_{g-1,g-1}^{(0)}(p-\hbar k, p-\hbar k) \\ + \frac{\Gamma\chi\chi'^* e^{i\delta t}}{\gamma^2 + \Delta^2} \int dq \bar{N}(q) \tilde{\rho}_{g1,g-1}^{(0)}(p+\hbar q, p+\hbar q) \\ - \frac{\chi\chi'^* e^{i\delta t}}{2(\gamma - i\Delta)} \tilde{\rho}_{g0,g0}^{(0)}(p+2\hbar k, p+2\hbar k) - \frac{\chi\chi'^* e^{i\delta t}}{2(\gamma + i\Delta)} \tilde{\rho}_{g0,g0}^{(0)}(p, p) \\ + \frac{\Gamma\chi\chi'^* e^{i\delta t}}{2(\gamma^2 - \Delta^2)} \int dq \bar{N}(q) \tilde{\rho}_{g0,g0}^{(0)}(p+\hbar k + \hbar q, p+\hbar k + \hbar q) \\ - \frac{\chi\chi'^* e^{i\delta t}}{6(\gamma - i\Delta)} \tilde{\rho}_{g1,g1}^{(0)}(p+3\hbar k, p+3\hbar k) - \frac{\chi\chi'^* e^{i\delta t}}{6(\gamma + i\Delta)} \tilde{\rho}_{g1,g1}^{(0)}(p+\hbar k, p+\hbar k)$$

$$\begin{aligned}
& + \frac{\Gamma \chi' e^{i\delta t}}{6(\gamma^2 + \Delta^2)} \int dq \tilde{N} \tilde{\rho}_{g-1,g1}^{(0)}(p + 2\hbar k + \hbar q, p + 2\hbar k + \hbar q) \\
& - \frac{\chi \chi'^* e^{i\delta t}}{6(\gamma + i\Delta')} \tilde{\rho}_{g-1,g1}^{(0)}(p - \hbar k, p + \hbar k) - \frac{\chi \chi'^* e^{i\delta t}}{6(\gamma - i\Delta)} \tilde{\rho}_{g-1,g1}^{(0)}(p + \hbar k, p + 3\hbar k) \\
& + \frac{\Gamma \chi' e^{i\delta t}}{6(\gamma^2 + \Delta^2)} \int dq \tilde{N}(q) \tilde{\rho}_{g-1,g1}^{(0)}(p + \hbar q, p + 2\hbar k + \hbar q). \tag{41}
\end{aligned}$$

It is shown in Appendix B that by imposing the following condition:

$$\frac{\Gamma'}{ku} \frac{\omega_k^2}{(ku)^2} \ll 1, \tag{42}$$

one can neglect the effects of the optical pumping terms in Eq. (41). Since the momentum diffusion coefficient is of order  $\Gamma' \hbar^2/k^2$ , and the atomic momentum distribution width is of order  $mu$ , the above condition implies that, during the time of order  $1/ku$  in which the ensemble of  $\tilde{\rho}_s^{(0)}(p, p + 2\hbar k)$  decays as a result of Doppler dephasing, the amount of momentum diffusion due to optical pumping is small compared with the initial momentum width  $mu$ . Assuming that condition (42) is satisfied, the terms on the right-hand side of Eq. (41) that are proportional to  $\Gamma_p$  can be dropped. Finally, we assume the zeroth-order terms in Eq. (41) are given by [3].

$$\begin{aligned}
\tilde{\rho}_{g-1,g+1}^{(0)}(p) &= \rho_{-1}^{(0)} W(p \mp \hbar k), \\
\tilde{\rho}_{g0,g0}^{(0)}(p) &= \rho_0^{(0)} W(p), \\
\tilde{\rho}_{g-1,g+1}^{(0)}(p) &= \rho_{-1,g+1}^{(0)} W(p), \tag{43}
\end{aligned}$$

where  $W(p)$  is a normalized distribution function of  $p$  which has a width of order  $p_0 = mu$  that satisfies inequality (27).

The population modulation term  $\rho_s(p)$  can now be solved from Eq. (41) without the optical pumping terms, and integrated over  $p$  to give the recoil-induced signal to lowest order in  $\omega_k/(ku)$  and  $\Gamma/|\Delta|$  as

$$\begin{aligned}
\rho_{ge}^{(re)} &= \frac{5}{9} \frac{i \chi' e^{-i\delta t}}{\gamma + i\Delta'} \int \rho_s(p) dp \\
&= -\frac{10}{9} \frac{\sqrt{\pi} \chi'^* |\chi|^2}{(\gamma + i\Delta') (\gamma^2 + \Delta^2)} \frac{\Delta \omega_k}{(ku)^2} I' \left[ \frac{i \gamma_g - \delta}{2ku} \right] \\
&\quad \times [\rho_{-1}^{(0)} + \frac{1}{2} \rho_0^{(0)} + \frac{1}{6} \rho_1^{(0)} + \frac{1}{6} \rho_{-1,1}^{(0)}]. \tag{44}
\end{aligned}$$

The function  $I(x)$  is defined as

$$I(x) = \frac{i}{\sqrt{\pi}} \int_x^\infty \frac{W(p_0 t) p_0 dt}{z - t}, \tag{45}$$

and  $I'(x)$  is the first-order derivative of  $I(x)$  with respect to  $x$ . The values of  $\rho_m^{(0)}$  can be deduced from the results of semiclassical calculations [3]. In the limit that  $ku \ll \Gamma'$ , they are given by

$$\rho_{-1}^{(0)} = \rho_{-1}^{(0)} = \frac{13}{14}, \quad \rho_0^{(0)} = \frac{4}{11}, \quad \rho_{-1,1}^{(0)} = \frac{5}{14}. \tag{46}$$

As shown in Ref. [2], the signal  $\text{Im}[\tilde{\rho}_{ge}^{rc}]$  as a function of  $\delta$  has a dispersion-like line shape around  $\delta = 0$  for a Gaussian distribution of  $W(p)$ , and it has the same sign as we have predicted qualitatively in the Introduction.

## B. Raman signal

We now consider the contribution to the probe absorption signal due to the atomic ground-state orientation and alignment, which can be interpreted as a Raman signal in the limit that  $|\Delta|/\Gamma \gg 1$ . Since both the ground-state energy splitting  $|\delta E| \sim |\chi|^2 |\Delta| / (\gamma^2 + \Delta^2)$  and the effective decay rate  $\Gamma'$  of each sublevel are much larger than  $\omega_k$ , one can neglect the recoil shift terms in considering the Raman signal. In this case, the calculation for the Raman signal can be carried out semiclassically in a sense that the atomic center-of-mass momentum and position are treated as classical variables. The semiclassical equations for the density matrix can be obtained directly from Eqs. (19)–(23) by the transformation [2]

$$\begin{aligned}
\tilde{\rho}_{gm,gm'}(z, t) &= \frac{1}{2\pi\hbar} \int \int dp dp' \exp \left[ i \frac{(p - p')z}{\hbar} \right] \\
&\quad \times \tilde{\rho}_{gm,gm'}(p, p'), \tag{47}
\end{aligned}$$

neglecting terms proportional to the recoil energy  $\hbar\omega_k$ . In Eq. (47),  $z$  is the classical atomic center-of-mass position, which can depend on time. Defining similar quantities for the atomic ground-state population, orientation and alignment as

$$\begin{aligned}
\rho_s &= \sum_{m=-1}^1 \tilde{\rho}_{gm,gm}(z, t), \\
\rho_{or} &= \frac{1}{\sqrt{2}} [\tilde{\rho}_{g1,g1}(z, t) - \tilde{\rho}_{g-1,g-1}(z, t)], \\
\rho_{al} &= \frac{1}{\sqrt{6}} [\tilde{\rho}_{g1,g1}(z, t) + \tilde{\rho}_{g-1,g-1}(z, t) \\
&\quad - 2\tilde{\rho}_{g0,g0}(z, t)], \\
\rho_{-1,1} &= \tilde{\rho}_{g-1,g+1}(z, t), \tag{48}
\end{aligned}$$

one then obtains the following equations:

$$\dot{\rho}_s = 0 , \quad (49)$$

$$\begin{aligned} \dot{\rho}_{\text{or}} &= -\frac{\Gamma'}{6}\rho_{\text{or}} + \frac{\Gamma'}{3\sqrt{2}}\frac{i\Delta}{\Gamma}(e^{-i2kz}\rho_{1,-1} - e^{i2kz}\rho_{-1,1}) \\ &\quad - \chi'^*\chi e^{-i2kz+i\delta t} \left| \frac{1}{\gamma-i\Delta} + \frac{1}{\gamma+i(\Delta+\delta)} \right| \left| \frac{5\sqrt{2}}{72}\rho_s + \frac{1}{12}\rho_{\text{or}} - \frac{\sqrt{3}}{18}\rho_{\text{al}} \right| \\ &\quad - \frac{1}{6\sqrt{2}}\chi'^*\chi e^{i\delta t} \left| \frac{1}{\gamma-i\Delta} - \frac{1}{\gamma+i(\Delta+\delta)} \right| \rho_{-1,1} , \end{aligned} \quad (50)$$

$$\begin{aligned} \dot{\rho}_{\text{al}} &= -\frac{11}{18}\Gamma' + \frac{5}{18\sqrt{6}}\Gamma' - \frac{\Gamma'}{3\sqrt{6}}(e^{-i2kz}\rho_{1,-1} + e^{i2kz}\rho_{-1,1}) \\ &\quad - \chi'^*\chi e^{-i2kz+i\delta t} \left| \frac{1}{\gamma-i\Delta} + \frac{1}{\gamma+i(\Delta+\delta)} \right| \left| -\frac{5}{36\sqrt{6}}\rho_s + \frac{1}{6\sqrt{3}}\rho_{\text{or}} + \frac{11}{36}\rho_{\text{al}} \right| \\ &\quad - \chi'^*\chi e^{i\delta t} \left| \frac{1}{\gamma-i\Delta} + \frac{1}{\gamma+i(\Delta+\delta)} \right| \left| \frac{1}{3\sqrt{6}}\rho_{-1,1} \right| , \end{aligned} \quad (51)$$

$$\begin{aligned} \dot{\rho}_{1,-1} &= -\frac{5}{6}\Gamma'\rho_{1,-1} + \frac{5}{36}\Gamma'e^{i2kz}\rho_s - \frac{\Gamma'}{3\sqrt{6}}e^{2ikz}\rho_{\text{al}} + \frac{\Gamma'}{3\sqrt{2}}\frac{i\Delta}{\Gamma}e^{i2kz}\rho_{\text{or}} \\ &\quad - \chi'^*\chi e^{i\delta t} \left| \frac{1}{\gamma-i\Delta} + \frac{1}{\gamma+i(\Delta+\delta)} \right| \left| -\frac{5}{36}\rho_s + \frac{1}{3\sqrt{6}}\rho_{\text{al}} \right| \\ &\quad + \chi'^*\chi e^{i\delta t} \left| \frac{1}{\gamma-i\Delta} - \frac{1}{\gamma+i(\Delta+\delta)} \right| \left| \frac{1}{6\sqrt{2}}\rho_{\text{or}} - \frac{5}{6}\frac{\chi'^*\chi}{\gamma+i(\Delta+\delta)}e^{-i2kz+i\delta t}\rho_{1,-1} \right| , \end{aligned} \quad (52)$$

$$\dot{\rho}_{-1,1} = -\frac{5}{6}\Gamma'\rho_{-1,1} + \frac{5}{36}\Gamma'e^{-i2kz}\rho_s - \frac{\Gamma'}{3\sqrt{6}}e^{-i2kz}\rho_{\text{al}} - \frac{\Gamma'}{3\sqrt{2}}\frac{i\Delta}{\Gamma}e^{-i2kz}\rho_{\text{or}} - \frac{5}{6}\frac{\chi'^*\chi}{\gamma-i\Delta}e^{-i2kz+i\delta t}\rho_{-1,1} . \quad (53)$$

As is evident from Eq. (49), the total population in the semiclassical picture is a conserved quantity, i.e.,

$$\rho_s = 1 . \quad (54)$$

We now make the following expansions in powers of the probe-field strength for the steady-state solutions of Eqs. (50)–(53):

$$\begin{aligned} \rho_{\text{or}} &= \rho_{\text{or}}^{(0)} + \rho_{\text{or}}^{(1)}e^{-i2kz+i\delta t} , \\ \rho_{\text{al}} &= \rho_{\text{al}}^{(0)} + \rho_{\text{al}}^{(1)}e^{-i2kz+i\delta t} , \\ \rho_{1,-1} &= \rho_{1,-1}^{(0)}e^{i2kz} + \rho_{1,-1}^{(1)}e^{i\delta t} , \\ \rho_{-1,1} &= \rho_{-1,1}^{(0)}e^{-i2kz} + \rho_{-1,1}^{(1)}e^{-i4kz+i\delta t} , \end{aligned} \quad (55)$$

where the superscript (0) or (1) again indicates that the related terms are to zeroth or to first order in probe-field strength. Equations (50)–(53) can then be solved recursively. First, the zeroth-order terms are solved by letting  $\chi' = 0$  in Eqs. (50)–(53). One finds

$$\begin{aligned} \rho_{\text{or}}^{(0)} &= \frac{100\sqrt{2}}{17} \frac{(\Delta/\Gamma)(kv/\Gamma')}{5+4(\Delta/\Gamma)^2 + \left| \frac{4400}{51} \right| (kv/\Gamma')^2} , \\ \rho_{\text{al}}^{(0)} &= \frac{5\sqrt{6}}{102} \left| 1 - \frac{800}{17} \frac{(kv/\Gamma')^2}{5+4(\Delta/\Gamma)^2 + \left| \frac{4400}{51} \right| (kv/\Gamma')^2} \right| , \\ \rho_{1,-1}^{(0)} &= \frac{5}{34} \frac{5+4(\Delta/\Gamma)^2 - i20(kv/\Gamma')}{5+4(\Delta/\Gamma)^2 + \left| \frac{4400}{51} \right| (kv/\Gamma')^2} , \\ \rho_{-1,1}^{(0)} &= [\rho_{1,-1}^{(0)}]^* , \end{aligned} \quad (56)$$

where  $v$  is the  $z$  component of the atomic velocity. One can verify that to zeroth order in  $kv/\Gamma'$ , Eqs. (54) and (56) lead to the results of Eq. (46) in Sec. III A.

Upon substitution of Eq. (56) into Eqs. (50)–(53), one can then solve for the ground-state elements to first order in probe-field strength. In the limit that  $ku/\Gamma' \sim 0$ , the first-order terms are given by

$$\begin{aligned} \rho_{\text{or}}^{(1)} &= -\frac{5\sqrt{2}}{204} \frac{\chi'^*\chi}{\Gamma'/6+i\delta+(\Gamma'/3)^2(\Delta/\Gamma)^2/(5\Gamma'/6+i\delta)} \\ &\quad \times \left| \left| 3 - \frac{5\Gamma'/3}{5\Gamma'/6+i\delta} \frac{i\Delta}{\Gamma} \right| \frac{1}{\gamma-i\Delta} \right. \\ &\quad \left. + \frac{2}{\gamma+i(\Delta+\delta)} \right| , \end{aligned}$$

$$\begin{aligned} \rho_{\text{al}}^{(1)} &= 0 , \\ \rho_{1,-1}^{(1)} &= -\frac{1}{3\sqrt{2}}\frac{i\Delta}{\Gamma}\frac{\Gamma'}{5\Gamma'/6+i\delta}\rho_{\text{or}}^{(1)} \\ &\quad + \frac{25}{204}\frac{1}{5\Gamma'/6+i\delta}\frac{\chi'^*\chi}{\gamma-i\Delta} , \\ \rho_{-1,1}^{(1)} &= -\rho_{1,-1}^{(1)*} . \end{aligned} \quad (57)$$

The Raman signal [see Eq. (14)] is a function of

$$\rho_{\text{re}}^{(\text{Rm})} = -\frac{5\sqrt{2}}{12}\frac{i\chi'^*\chi}{\gamma+i\Delta} \left| \rho_{\text{or}}^{(1)} - \frac{1}{5\sqrt{3}}\rho_{\text{al}}^{(1)} - \frac{\sqrt{2}}{5}\rho_{1,-1}^{(1)} \right| . \quad (58)$$

Upon substitution of the results (57) into Eq. (58), one obtains the Raman signal to zeroth order in  $ku/\Gamma'$  as

$$\tilde{\rho}_{ge}^{(Rm)} = \frac{25}{1224} \frac{i\chi'^* |\chi|^2}{\gamma + i\Delta} \left| \frac{1 - \frac{\Gamma'/15}{5\Gamma/6 + i\delta} \frac{i\Delta}{\Gamma}}{\Gamma'/6 + i\delta + \frac{(\Gamma'/3)^2 (\Delta/\Gamma)^2}{5\Gamma'/6 + i\delta}} \right| \left| 3 - \frac{5\Gamma'/3}{5\Gamma'/6 + i\delta} \frac{i\Delta}{\Gamma} + \frac{2}{\gamma - i\Delta} \right| + \frac{1}{5\Gamma'/6 + i\delta} \frac{1}{\gamma - i\Delta} \right|. \quad (59)$$

In the limit  $|\Delta| \gg \Gamma$ , the Raman signal, proportional to  $\text{Im}[\tilde{\rho}_{ge}^{(Rm)}]$ , exhibits resonances at

$$\delta = \pm \frac{\Gamma' \Delta}{3 \Gamma}. \quad (60)$$

Both resonances are Lorentzian type with opposite signs, and the full width at half maximum (FWHM) of each resonance is given by  $\Gamma'$ . For negative  $\Delta$ , the probe gain and probe absorption occur when  $\delta$  equals  $\Gamma'\Delta/(3\Gamma)$  and  $-\Gamma'\Delta/(3\Gamma)$ , respectively. The ratio between the magnitudes of the gain and the absorption peaks is  $\frac{4}{3}$ .

The above result can be understood if one chooses the quantization axis along the local direction of the total pump-field polarization. As shown in Ref. [3], the combination of the counter-propagating  $\sigma^+ - \sigma^-$  pump field results in a linearly polarized field whose strength is space independent and whose polarization vector rotates along the laser propagation axis. Relative to the local quantization axis, the pump field is  $\pi$  polarized with an amplitude equal to  $\sqrt{2}\delta$ , while the probe polarization has  $\sigma^+$ ,  $\sigma^-$ , and  $\pi$  components. Owing to different Clebsch-Gordan coefficients for different transitions (see Fig. 2), to zeroth order in probe-field strength, the steady-state population of the  $m=0$  state is greater than those of the  $m=\pm 1$  states, and the light shifts of these states induced by the pump field also differ. One can verify easily that the energy splitting between the  $m=0$  and  $\pm 1$  states  $\delta_{0,\pm 1}$  is given by Eq. (60). For  $|\Delta| \gg \Gamma$  and negative  $\Delta$ , when  $\delta < 0$ , the dominant process is the transition from  $m=0$  to  $\pm 1$  states via absorption of a pump photon and emission of a probe photon, resulting in probe gain; when  $\delta > 0$ , the same transition involves absorption of a probe photon and emission of a pump photon, resulting in probe loss. As shown in Fig. 2(a), these two processes have relative strength  $[(1/\sqrt{6})\sqrt{2}/3]^2 / [(1/\sqrt{2})(1/\sqrt{2})]^2 = \frac{4}{9}$ .

### C. Overall signal

The overall signal is proportional to

$$\text{Im}(\tilde{\rho}_{ge}) = \text{Im}[\tilde{\rho}_{ge}^{(rc)}] + \text{Im}[\tilde{\rho}_{ge}^{(Rm)}], \quad (61)$$

which is a superposition of the recoil and Raman signals. When  $|\Delta| \gg \gamma$ , both signals are proportional to  $1/|\Delta|^2$ . The dimensionless parameter that determines the magnitude of the recoil signal to that of the Raman signal is given by

$$\eta = \frac{\omega_k \Gamma'}{\xi (ku)^2}, \quad (62)$$

where  $\xi < 1$  is a parameter that characterizes the popula-

tion difference between the initial and final states of the Raman transition.

Figures 3(a) and 3(b) show the probe absorption signal as a function of  $\delta$  for two different sets of parameters, with different values of  $\Delta/\gamma$ . The recoil-induced signal is centered at  $\delta=0$ , having width of order  $2ku$ . The Raman signal has a width equal to  $\Gamma'$ . The distance between the two Raman peaks corresponds to the differential light shifts between different magnetic substates. As one can see from Fig. 3, when  $|\Delta|/\gamma$  increases, the splitting of the two Raman peaks also increases, while the recoil-induced signal remains a dispersionlike curve.

For ideal sub-Doppler cooling, one can utilize a relation

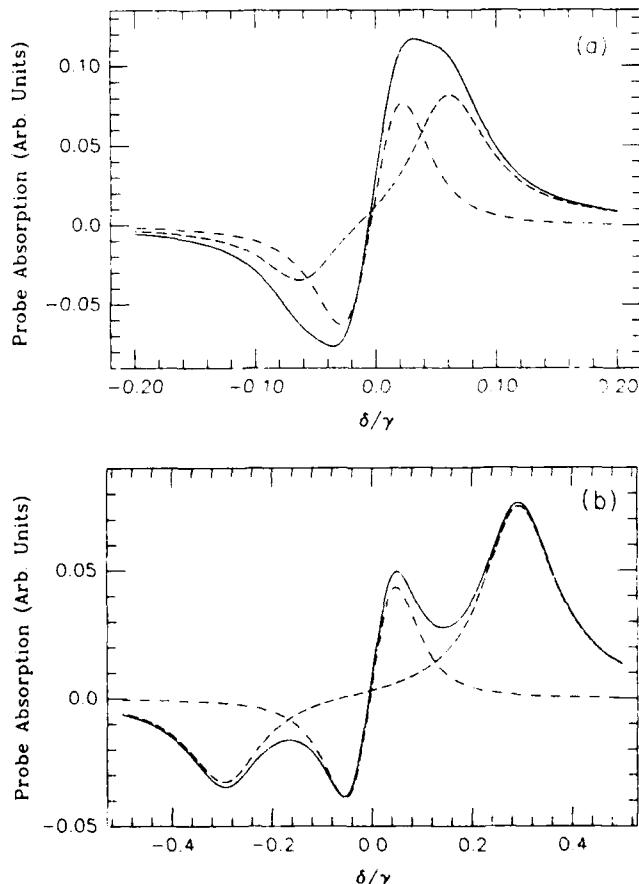


FIG. 3. Probe absorption coefficient. The dashed line shows the contribution of the Raman resonances, while the dot-dashed line shows that of the recoil-induced resonances. The solid line shows the overall signal. For both figures,  $\omega_k/\gamma = 0.0008$ , and  $\gamma_k/\gamma = 0.0001$ . (a)  $\Delta/\gamma = -5.0$ ,  $\chi/\gamma = 1.0$ ,  $ku/\gamma = 0.02$ ; (b)  $\Delta/\gamma = -10.0$ ,  $\chi/\gamma = 3.0$ ,  $ku/\gamma = 0.04$ .

$$\frac{\omega_k}{(ku)^2} \sim \frac{\hbar}{k_B T} \sim \frac{|\Delta|}{|\chi|^2}, \quad (63)$$

to rewrite  $\eta$  as

$$\eta \sim \frac{1}{\xi} \frac{\gamma}{|\Delta|}. \quad (64)$$

For values of  $|\Delta|/\gamma$  that are of order  $1/\xi$ , the magnitude of the recoil-induced probe absorption signal is comparable to that of the Raman signal. Equation (63) also indicates that the width of the recoil-induced signal, which is of order  $ku$ , increases linearly with  $|\chi|$  for a fixed value of  $|\Delta|$ . This is in contrast with the Raman signal, whose width [ $\sim \Gamma'$ ] increases quadratically with  $|\chi|$ . Finally, when the cooling condition is not optimal and  $ku$  is of order  $\Gamma'$ , the Raman signal will be a convolution of a Lorentzian function of width  $\Gamma'$  with a Gaussian function of width  $1.66(2ku)$ .

#### IV. DISCUSSION

We have presented the calculations of probe absorption coefficients in the  $J_g=1 \rightarrow J_e=2$  systems in the presence of polarization gradient pump fields. Apart from the Raman signals which result from the nonconservation of the atomic properties such as the ground-state orientation or alignment, there is a recoil-induced signal which is related to a probe-induced spatial modulation of the atomic total population. We find that optical pumping does not broaden the recoil-induced resonances. This can be understood as follows. In the picture of Raman-type processes between center-of-mass momentum states, the transition width is determined by the effective lifetime of each momentum subclass. When condition (42) is satisfied [ $(\Gamma'/ku)(\omega_k^2/k^2 u^2) \ll 1$ ], the population in a given momentum interval  $\Delta p$  does not decay during the time interval  $1/(ku)$ . As a result the optical pumping does not broaden the linewidth of the recoil-induced signal.

The recoil-induced signal calculated in Sec. III is related to the stimulated transitions between different momentum families involving the pump and the probe fields, owing to this particular probe configuration. On the other hand, if the probe is a  $\sigma^+$  field copropagating with the  $\sigma^+$  pump, the stimulated transitions involving the probe field are between states that belong to the same family. As a result, there is no recoil-induced signal in this case. However, it has been shown recently by Lounis *et al.* [9] that there is a Rayleigh-type resonance in this situation even without the inclusion of recoil effects, owing to some nonvanishing spatially averaged atomic velocity that oscillates at a frequency  $\delta$  under the influence of the copropagating pump and probe fields having the same polarization. This average velocity leads to a so-called motion-induced atomic orientation [3], which scatters photons from the pump field into the probe field, resulting in the probe gain or loss. Such a Rayleigh resonance will not be broadened by the atomic Doppler width since it only involves transitions between atomic states of the same momentum. It is clear from the above arguments that there is no Rayleigh resonance for the field configuration

considered here, since the spatially averaged atomic velocity, to first order in the probe-field strength, is zero. As a result there is no motion-induced atomic orientation in this case. Instead, there are recoil-induced resonances, i.e., Raman processes between atomic states of different kinetic energies, for this field geometry. The width of the recoil-induced signal is determined by the Doppler width  $2ku$  and serves as a probe of the atomic velocity distribution.

The theoretical calculations presented here cannot be compared directly with the experimental results on pump-probe spectroscopy of atoms confined in a magneto-optical trap [4]. One reason is that the actual atomic level schemes are more complicated ( $F=4 \rightarrow F=5$  for Cs atoms) than that considered in this work. Also the possible inclusion of inhomogeneous magnetic fields in those earlier works can further complicate the problem. In the results of some more recent experiments in one-dimensional (1D) optical molasses produced by a pair of circularly polarized fields [9,10], there have been some signatures of the recoil-induced resonances considered in this and a previous paper [2]. However, a direct, unambiguous observation of such resonance phenomena is yet to be made. When selecting the field configuration as the one considered here, it may be useful to choose parameters such that  $|\Delta|/\Gamma' \gg 1$ , thereby the normal Raman signals can be separated from the recoil-induced signal centered at  $\delta=0$ .

Finally, we discuss the relation between the results calculated here, and a number of experiments on cold atoms using lin||lin cooling field configuration [11,12], in which the linear polarizations of the counterpropagating cooling fields are orthogonal. As is well known, atoms in such field configurations experience an effective, spatially modulated potential originating from the atomic ground-state energy shifts in the fields. If the average atomic kinetic energy is cooled below such potential depth, a significant portion of the atomic population becomes spatially localized [13]. Such localized atoms have a bandlike energy structure [14], which can be probed by various spectroscopic means. In situations like this, where spatial localization of atoms is important, the recoil-induced resonances, which are basically a phenomenon associated with the atoms in the energy continuum states, may still exist. Due to the small population of untrapped atoms in most situations, and also possibly due to the large amplitude of the signal provided by the trapped atoms [15], the recoil-induced signal may be too small to be observable under most experimental conditions. There might be some cases, for example, with low cooling field intensities, where the localization effects are less significant, and the recoil-related effects are more pronounced. Whether such situations exist requires further research.

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## APPENDIX A

In this Appendix, we give the derivation of the probe absorption coefficients in terms of the atomic ground-state density-matrix elements. A convenient basis for the derivation is the irreducible tensor basis [5,16], in which density-matrix elements are defined as

$$\rho_Q^k(F_1, F_2) = \sum_{m_1, m_2} (-1)^{F_2 - m_2} \langle F_1, m_1; F_2, -m_2 | K, Q \rangle \\ \times \rho_{F_1, m_1; F_2, m_2}, \quad (\text{A1})$$

where  $\rho_{F_1, m_1; F_2, m_2}$  are density-matrix elements written in the magnetic-state basis, and  $\langle F_1, m_1; F_2, -m_2 | K, Q \rangle$  is a Clebsch-Gordan coefficient. The polarization of the medium can be written as [16]

$$\mathbf{P} = \frac{1}{\sqrt{3}} (-1)^{H-G+1} e r_{eg} \sum_{q=\pm 1,0} \epsilon_q \rho_q^1(G, H) + \text{c.c.}, \quad (\text{A2})$$

where  $G, H$  are the angular momenta of the ground and excited states, respectively,  $r_{eg}$  is a reduced matrix element of the dipole moment, and

$$\epsilon_{\pm 1} = \mp \frac{1}{\sqrt{2}} (\hat{x} \pm i \hat{y}), \quad \epsilon_0 = \hat{z}. \quad (\text{A3})$$

The absorption coefficient for a probe field having (complex) polarization  $\epsilon'$ , frequency  $\Omega'$ , and propagation vector  $\mathbf{k}'$ , is proportional to [17]

$$\epsilon'^* \cdot \mathbf{P}(\mathbf{k}') = \sum_q (-1)^q \epsilon'_q P_{-q}(\mathbf{k}') \\ = \frac{e r_{eg}}{\sqrt{3}} (-1)^{H-G+1} \sum_q \epsilon'_q \rho_q^1(G, H; \mathbf{k}'), \quad (\text{A4})$$

where  $\mathbf{P}(\mathbf{k}')$  and  $\rho_q^1(G, H; \mathbf{k}')$  denote the components of  $\mathbf{P}$  and  $\rho_q^1(G, H)$  that vary as  $\exp(-i\mathbf{k}' \cdot \mathbf{R} + i\Omega' t)$ , and the  $\epsilon'_q$ 's are given by

$$\epsilon'_{\pm 1} = \mp \frac{1}{\sqrt{2}} (\epsilon'_x \pm i \epsilon'_y), \quad \epsilon'_0 = \epsilon'_z, \quad (\text{A5})$$

where  $\epsilon'_x$ ,  $\epsilon'_y$ , and  $\epsilon'_z$  are the Cartesian components of the probe-field polarization vector.

In the case of Sec. III, the probe polarization is given by

$$\epsilon_q = -\delta_{q,1}. \quad (\text{A6})$$

Substituting Eq. (A6) into Eq. (A4), one obtains

$$\tilde{\rho}_{ge} \propto \tilde{\rho}_1^1(G, H; k), \quad (\text{A7})$$

where

$$\tilde{\rho}_1^1(G, H; k) = \rho_1^1(G, H; k) e^{ikz - i\omega' t}. \quad (\text{A8})$$

In the magnetic-state basis, one has

$$\tilde{\rho}_{ge} = \left[ \tilde{\rho}_{g-1,e-2}(k) + \frac{1}{\sqrt{2}} \tilde{\rho}_{g0,e-1}(k) \right. \\ \left. + \frac{1}{\sqrt{6}} \tilde{\rho}_{g1,e0}(k) \right] e^{-i\Delta't}, \quad (\text{A9})$$

where  $\tilde{\rho}_{gm,em}(k)$  are related to density-matrix elements  $\tilde{\rho}_{gm,em}(p, p')$  defined in Eq. (9) through

$$\tilde{\rho}_{gm,em}(k) = \int \tilde{\rho}_{gm,em}(p, p + \hbar k) dp. \quad (\text{A10})$$

When replacing  $\tilde{\rho}_{gm,em}$  with ground-state matrix elements, and neglecting terms that correspond to linear probe absorption, one obtains the following expression for  $\tilde{\rho}_{ge}$ :

$$\tilde{\rho}_{ge} = \frac{i \chi_-^* e^{-i\delta t}}{\gamma + i\Delta'} \int [\tilde{\rho}_{-1}^{(1)}(p, p + 2\hbar k) + \frac{1}{2} \tilde{\rho}_0^{(1)}(p, p + 2\hbar k) \\ + \frac{1}{6} \tilde{\rho}_1^{(1)}(p, p + 2\hbar k)] dp \\ + \frac{i \chi_+^* e^{-i\delta t}}{6(\gamma + i\Delta')} \int \tilde{\rho}_{g1,g-1}^{(1)}(p, p) dp, \quad (\text{A11})$$

where  $\tilde{\rho}_{\pm 1}^{(1)}$  and  $\tilde{\rho}_0^{(1)}$  denote  $\tilde{\rho}_{g\pm 1,g\pm 1}^{(1)}$  and  $\tilde{\rho}_{g0,g0}^{(1)}$  respectively. By using the identity

$$\tilde{\rho}_{-1} + \frac{1}{2} \tilde{\rho}_0 + \frac{1}{6} \tilde{\rho}_1 = \frac{5}{9} \tilde{\rho}_s - \frac{5}{12} (\tilde{\rho}_1 - \tilde{\rho}_{-1}) \\ + \frac{1}{36} (\tilde{\rho}_1 + \tilde{\rho}_{-1} - 2\tilde{\rho}_0), \quad (\text{A12})$$

where  $\tilde{\rho}_s$  is the total population as defined in Eq. (16), one obtains Eq. (14) for  $\tilde{\rho}_{ge}$ .

## APPENDIX B

In the appendix, we analyze the effects of optical pumping due to the presence of the pump fields on the recoil-induced signal, and give the derivation of condition (42). The optical pumping processes cause an effective atomic momentum diffusion, which can be represented by Eq. (40). Instead of using an effective transit decay rate  $\gamma_g$ , we solve the time-dependent problem for the recoil signal. The evolution equation of  $\rho(p)$  can be written from Eq. (41) as

$$\dot{\rho}_s(p) = -2\Gamma_p \rho_s(p) + 2\Gamma_p \int \omega(p' \rightarrow p) \rho_s(p') dp' \\ + i \left[ \frac{2kp}{m} + 4\omega_k \right] \rho(p) + e^{i\delta t} W_0(p), \quad (\text{B1})$$

where  $W_0(p)$  represents the “source” terms in Eq. (41) (terms explicitly proportional to  $\chi^{**}$ ). The kernel  $\omega(p' \rightarrow p)$  is given by

$$\omega(p' \rightarrow p) = \frac{1}{2} [\delta(p' - p + \hbar k) + \delta(p' - p - \hbar k)]. \quad (\text{B2})$$

Equation (B1) can be solved in the interaction representation defined by

$$\rho_s(p) = \tilde{\rho}(p) e^{i(2kp/m + 4\omega_k)t}. \quad (\text{B3})$$

Moreover, by noting that  $\omega(p' \rightarrow p)$  depends only on the difference  $(p - p')$ , one can make a Fourier transformation,

$$\bar{\rho}(p) = \int \rho(\zeta) e^{i\zeta p} d\zeta, \quad (B4)$$

and obtain the solution for  $\rho(\zeta)$  as follows:

$$\begin{aligned} \rho(\zeta) = & \exp[-2\Gamma_p t + 2\Gamma_p \int_0^t \tilde{\omega}(\zeta, t') dt'] \\ & \times \int_0^t \exp[2\Gamma_g t' - 2\Gamma_p \int_0^{t''} \tilde{\omega}(\zeta, t'') dt''] \tilde{\omega}_s(\zeta, t') dt', \end{aligned} \quad (B5)$$

where

$$\begin{aligned} \tilde{\omega}(\zeta, t) = & \frac{1}{2} \{ \exp[-i\hbar\zeta k - i4\omega_k t] \\ & + \exp[i\hbar\zeta k + i4\omega_k t] \}, \end{aligned} \quad (B6)$$

and

$$\tilde{\omega}_s(\zeta, t) = \int W_0(p) e^{i\delta t - i(2kp/m)t - i4\omega_k t} e^{-i\zeta p} dp. \quad (B7)$$

The recoil related probe absorption signal depends on  $\int e^{i\delta t} \rho(p)_s dp$  [see Eqs. (14), (15), (30), and (32)]. From Eqs. (B3) and (B4), it's easy to show that

$$\int e^{-i\delta t} \rho(p)_s dp = \hbar \left[ \frac{-2k}{m} t \right] e^{-i\delta t + i4\omega_k t}. \quad (B8)$$

As an illustration, let us assume  $W_0(p)$  to be a Gaussian

centered at  $p=0$ , with the most probable momentum  $p_0=mu$ . Then upon substitution of Eqs. (B5)–(B7) into Eq. (B8), one obtains

$$\begin{aligned} \int \rho(p) dp = & \int_0' \exp \left[ -2\Gamma_p t' + 2\Gamma_p \frac{\sin(4\omega_k t')}{4\omega_k} \right] \\ & \times \exp[-(kut')^2 - i\delta t' + i4\omega_k t'] dt'. \end{aligned} \quad (B9)$$

It is obvious that the values of  $t'$  that contribute to the above integral are  $t' \leq 1/(ku)$ . All the effects of the optical pumping are included in the first exponential term on the right-hand side of Eq. (B9). In order for the pumping effects to be negligible, this term has to be approximately unity. Since  $\omega_k/(ku) \ll 1$ , one can expand the sine function and reach the following expression:

$$\exp \left[ -\frac{4}{3} \frac{\Gamma_p}{ku} \frac{\hbar^2 k^2}{m^2 u^2} \right] \approx 1, \quad (B10)$$

which is true when condition (42) is satisfied. The momentum diffusion coefficient due to the optical pumping effects is  $\Gamma_p \hbar^2 k^2$ . Condition (42) implies that, during the coherence dephasing time, the amount of momentum diffusion due to optical pumping is small compared with the width of the original momentum distribution.

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